

REMARKS

The Official Action dated March 26, 2008 has been carefully considered. Accordingly, the present Amendment is believed sufficient to place the present application in condition for allowance. Reconsideration is respectfully requested.

By the present amendment, claim 1 has been amended to include limitations from claim 4 and to more clearly recite that the step of calculating the amount of hydrogen sulfide discharged from the reaction vessel uses a predetermined relational expression, in accordance with the teachings in the specification, for example at pages 24-25 and 32-33. Claims 1, 2 and 8 are also amended for matters of form only. Since these changes are fully supported by the original specification, entry of the present Amendment is warranted and respectfully requested.

In the Official Action, claims 1-9 were rejected under 35 U.S.C. §102(b) as anticipated by, or under 35 U.S.C. §103(a) as obvious over, the Miyahara et al U.S. Patent No. 5,840,830.

However, as will be set forth in detail below, Applicants submit that the processes for producing a poly(arylene sulfide) defined by the present claims 1-3 and 5-9 are not anticipated by and are non-obvious over Miyahara et al. Accordingly, this rejection is traversed and reconsideration is respectfully requested.

It is an object of the present invention to provide a reliable process for producing good-quality poly(arylene sulfides) (hereinafter abbreviated as "PASs"). In order to do so, it is necessary to strictly control various polymerization conditions, including the molar ratio of alkali metal sulfide to dihalo-aromatic compound, water content, polymerization temperature, and polymerization time, even when the purities of raw materials, secondary raw materials and the like are high and constant.

The hydrate of an alkali metal sulfide is generally used as a raw material in the polymerization process. The hydrate contains a high amount of water of crystallization. Further, the hydrate and/or other raw materials may be added to the reaction system as aqueous solutions. Accordingly, a high level of water exists in the system once the raw materials have been charged into the reaction vessel. Thus, a dehydration step is generally employed prior to the polymerization step to control the water content in the PAS polymerization reaction system. In the dehydration step, the alkali metal sulfide reacts with water in the organic amide solvent, and hydrogen sulfide (H_2S) is equilibriously dissociated and volatilized. However, volatilization of hydrogen sulfide in the dehydration step causes the several problems in the industrial production of PAS.

First, the amount of the sulfur source in the reaction system after dehydration will vary according to the volatilization of hydrogen sulfide; as a result, the melt viscosities of the resulting polymers will vary by each lot. Second, it is difficult to stably produce a PAS having a high polymerization degree due to the volatilization of hydrogen sulfide. That is, since the polymerization reaction between the alkali metal sulfide (i.e., the sulfur source) and the dihalo-aromatic compound is a polycondensation reaction, it is desirable to closely regulate the molar ratio between these components to about 1:1 in order to provide a PAS having a high polymerization degree. Thus, attempts have been made to predict the amount of hydrogen sulfide volatilized in the dehydration step in order to adjust the amount of the sulfur source (alkali metal sulfide and/or alkali metal hydrosulfide) which is charged and control the sulfur source: dihalo-aromatic molar ratio. However, it is difficult to accurately control the molar ratio between both components in the polymerization reaction because the amount of hydrogen sulfide volatilized during dehydration varies widely, and, if the amount of hydrogen sulfide actually

volatilized out is less than the predicted amount, the molar ratio of the alkali metal sulfide to the dihalo-aromatic compound is excessive and undesirable side reactions such as rapid decomposition reaction tend to occur.

Some proposals have heretofore been made for the purpose of solving the problems caused by volatilization of hydrogen sulfide in the dehydration step. For example, one proposed process comprises determining the amount of hydrogen sulfide volatilized in a dehydration step to determine the amount of a sulfur source existing in the reaction system with high accuracy. According to this process, a molar ratio of the alkali metal sulfide to the dihalo-aromatic compound in the polymerization step may be controlled with high accuracy. However, it is necessary to introduce a special dedicated device for the determination of the amount of hydrogen sulfide volatilized in the dehydration step, and the quantitative analysis requires a significant amount of time, thereby increasing product costs.

The present invention overcome these various disadvantages and provides a production process by which a PAS having a desired melt viscosity can be reliably produced. In particular, the amount of hydrogen sulfide lost by volatilization out of the reaction system is accurately calculated by a comparatively simple method, whereby the molar ratio of the sulfur source charged to the dihalo-aromatic compound charged is accurately controlled, thereby reliably producing a PAS having a desired melt viscosity.

Particularly, the inventors have conceived of a method for calculating an amount of hydrogen sulfide discharged from a reaction vessel in a dehydration step prior to the polymerization step. Specifically, with reference to claim 1, the dehydration step is performed by a process comprising (i) heating the mixture in the reaction vessel, to which a distillation column is linked, and guiding volatilized vapor to the distillation column to distill and separate it

into respective components, (ii) refluxing a high-boiling fraction taken out of the bottom of the distillation column and comprising the organic amide solvent as a principal component into the reaction vessel, (iii) cooling a low-boiling fraction taken out of the top of the distillation column and comprising water and hydrogen sulfide on the other hand to discharge hydrogen sulfide that is not condensed by the cooling and reflux a part of the condensed water into the distillation column, and (iv) discharging the remaining water. A predetermined relational expression between the total of the amount of water refluxed into the distillation column and the amount of water discharged without being refluxed, and the amount of hydrogen sulfide discharged from the reaction vessel is used to calculate the amount of discharged hydrogen sulfide from the measured total amount of water. Such a relational expression is obtained by carrying out regression analysis using voluminous experimental data actually measured as to the relation between the total amount of water and the amount of hydrogen sulfide volatilized out in the dehydration step as a database. When the database is subjected to regression analysis, a relational expression of a linear model or non-linear model (double logarithm model or semilogarithm model) can be prepared. The amount of the sulfur source remaining in the mixture after the dehydration step (the charged sulfur source) is then calculated on the basis of the calculated amount of discharged hydrogen sulfide. A charged molar ratio of the sulfur source to the dihalo-aromatic compound is then controlled on the basis of the amount of the calculated charged sulfur source.

According to this method, the amount of discharged hydrogen sulfide can be accurately calculated without collecting discharged hydrogen sulfide and conducting quantitative analysis, and PASs with extremely consistent melt viscosity may be reliably produced.

Fig. 1 of the present application illustrates an exemplary apparatus used in the production process according to the present invention. An upper part of a reaction vessel 1 for example, a polymerization vessel, in which a dehydration reaction is performed, is connected to a distillation column 3 through a line 2 and successively connected to a condenser 5 and a storage tank 6 through a line 4 extending from the top of the distillation column 3. The distillation column 3 is, for example, a high performance distillation column capable of efficiently separating an organic amide solvent such as NMP (N-methyl-2-pyrrolidone) and water. In the method according to the present invention, a part of the water stored in the storage tank 6 is refluxed into the distillation column 3, integrated by a flowmeter 9. Refluxing a part of the water within the storage tank 6 into the distillation column 3 permits smooth and efficient separation between the organic amide solvent and water by the distillation column 3. In general, the water is desirably refluxed into the top or upper part of the distillation column.

In the dehydration step of claim 1, a weight ratio of an amount of water refluxed into the distillation column to an amount of water discharged without being refluxed is within a range of from 90:10 to 10:90. Assuming that the amount of water refluxed into the distillation column is R, and the amount of water discharged without being refluxed is E, a higher R/E ratio causes an increase in the load upon heating in the distillation column, resulting in energy loss. On the other hand, if the ratio R/E is lower, separation between the organic amide solvent and water within the distillation column is decreased and a part of the organic amide solvent is distilled out together with water from the top of the distillation column and discharged, which causes handling problems and loss of the organic amide solvent.

In the present invention, thus, an amount (that is, the actual “amount of the sulfur source charged”) of the sulfur source remaining in the mixture after the dehydration step is calculated

on the basis of the calculated amount of discharged hydrogen sulfide. The charged molar ratio between the sulfur source (A) and the dihalo-aromatic compound (B) is then controlled on the basis of the amount of the calculated sulfur source (A).

Accordingly, in the present production process of PAS comprising the polymerization of a sulfur source and a dihalo-aromatic compound in an organic amide solvent, the prior dehydration step can be smoothly performed and a molar ratio of the sulfur source charged to the dihalo-aromatic compound charged can be accurately controlled with the relational expression between the volatilized hydrogen sulfide and the amount of distillate of water upon distillation, thereby stably producing a PAS having a desired melt viscosity. In this regard, the Examiner's attention is directed to the resulting data from the examples and comparisons which are summarized in Tables 1 and 2 of the specification. Surprisingly, the results set forth in Table 1 demonstrate that PASs having a desired melt viscosity can be reliably and consistently produced according to the production process of the present invention and the product polymers exhibit little variation of melt viscosity by lot.

On the other hand, when the amount of hydrogen sulfide volatilized out in the dehydration step is determined from conventional methods (Comparative Examples 1 to 10), the melt viscosities of the product polymers vary greatly by lot, even with little change in conditions of the dehydration step as shown in Table 2. On the other hand, when an amount of hydrogen sulfide volatilized out in the dehydration step is measured (Comparative Example 11), a PAS having a desired melt viscosity can be obtained. However, a special measuring device is required to determine the amount of hydrogen sulfide, and the time required to measure the amount increases production time and costs.

The Examiner asserted that Miyahara et al disclose a process for producing a poly(arylene sulfide) comprising a dehydration step and a subsequent polymerization step as recited in the preamble of claim 1. The Examiner stated that the disclosure of Miyahara et al differs from the claims in that it does not disclose (1) the step of refluxing a fraction taken out of the bottom of the distillation column, (2) the step of determining a relational expression between the total amount of water refluxed into the distillation column and an amount of water discharged without being refluxed, (3) the amount of hydrogen sulfide discharged from the reaction vessel in advance, and (4) the claimed prepolymer's melt viscosity (as in claim 9). The Examiner asserted, however, that Miyahara et al disclose the use of a distillation column (column 6, line 45) wherein the water content can be controlled and the hydrogen sulfide is recovered and reused in the polymerization reaction, encompassing the claimed steps of the process. The Examiner also asserted it would therefore be inherent that a prepolymer having the claimed characteristic of melt viscosity could be prepared following the teachings of Miyahara et al.

However, Applicants submit that Miyahara et al neither anticipate nor render obvious the presently claimed process. Specifically, the process of Miyahara et al is discussed in the present specification as follows:

“There has been proposed a process comprising recovering hydrogen sulfide volatilized out during a dehydration step by causing it to be absorbed in an organic amide solvent outside the system, in which the dehydration step is being conducted, and reusing the hydrogen sulfide recovered as a sulfur source in a polymerization reaction (for example, Japanese Patent Application Laid-Open No. 286861/1997). According to this process, the hydrogen sulfide volatilized out in the dehydration step can be recovered and reused, thereby solving various problems attending on the volatilization of hydrogen sulfide and producing a PAS little in variation of melt viscosity and stable in quality. However, even in this process, the amount of the volatilized hydrogen sulfide absorbed in the organic amide solvent varies, and there is a demand for developing a method for more accurately grasping the amount of hydrogen sulfide” (page 7, beginning at line 10).

The process of the present invention as defined by claim 1 differs from the teachings of Miyahara et al in many significant aspects. First, the production process according to the present invention comprises a dehydration step (1), in which volatilized vapor is guided to a linked distillation column to distill and separate the vapor into respective components, and a fraction taken out of the bottom of the distillation column and comprising the organic amide solvent as a principal component is refluxed into the reaction vessel.

On the other hand, in the production process of Miyahara et al, no reaction vessel to which the distillation column is linked, is used in the dehydration step. To the contrary, Miyahara et al disclose that the dehydration step is conducted by distillation outside the reaction system (column 6, lines 43-46). Further, in the production process of Miyahara et al, the organic amide fraction is neither separated from the vapor volatilized upon the dehydration under heat nor refluxed into the reaction vessel. Specifically, Example 1a of Miyahara et al describes that:

“A 20-liter autoclave was equipped with a Snyder’s column of 3 cm in diameter, 90 cm in length and 10 stages. Further, a condenser was connected to the upper part of the Snyder’s column so as to collect a distillate cooled by this condenser in a three-necked flask through a rubber tube (line). The three-necked flask was provided with a condenser and a gas line. The gas line from the three-necked flask was connected to gas absorber (A) charged with 541 g of NMP, and a gas line from the gas absorber (A) was connected to a gas absorber (B) charged with 500 g of a 10% aqueous solution of NaOH so as to finally discharge the gas passed through the gas absorber (B) into the air” (Miyahara et al, column 11, line 66 to column 12, line 10).

As described above, in the production process of Miyahara et al, vapor volatilized by the dehydration under heat is cooled by the condenser, and the cooled distillate is collected in the three-necked flask. In other words, in the production process of Miyahara et al, organic amide

fraction is neither separated from the vapor volatilized upon the dehydration under heat in the dehydration step nor refluxed into the reaction vessel (autoclave). The remaining Examples of Miyahara et al employ the same process as in Example 1a.

Second, in the production process according to the present invention, a fraction taken out of the top of the distillation column and comprising water and hydrogen sulfide is cooled in the dehydration step (1) to discharge hydrogen sulfide that is not condensed by the cooling and reflux a part of water condensed into the distillation column, and the remaining water is discharged, wherein a weight ratio of an amount of water refluxed into the distillation column to an amount of water discharged without being refluxed is within a range of from 90:10 to 10:90.

On the other hand, in the production process of Miyahara et al, a part of the condensed water is not refluxed into the distillation column in the cooling step. Miyahara et al describe that:

“When water is distilled off by the heating in the dehydration step, the water is usually discharged in the form of an azeotropic mixture with the organic amide solvent out of the system. Alternatively, the organic amide solvent and water are separated from each other by distillation, and only water is discharged” (Miyahara et al, column 2, lines 28-33).

In the production process of Example 1a of Miyahara et al, water volatilized out of the autoclave (reaction vessel) in the cooling step is collected in the three-necked flask but is not refluxed into the distillation column.

Third, in the production process according to the present invention, a predetermined relational expression between the total of the amount of water refluxed into the distillation column and the amount of water discharged without being refluxed, and the amount of hydrogen sulfide discharged from the reaction vessel is used to calculate an amount of hydrogen sulfide discharged from the reaction vessel in the dehydration step from a measured value of the total amount of water in the dehydration step. An amount of the sulfur source remaining in the

mixture after the dehydration step is then calculated on the basis of the calculated amount of discharged hydrogen sulfide and a charged molar ratio of the sulfur source to a dihalo-aromatic compound is controlled on the basis of the amount of the calculated sulfur source.

On the other hand, in the production process of Miyahara et al, hydrogen sulfide volatilized out in the dehydration step is absorbed and collected in an organic amide solvent outside the system in which the dehydration step is being conducted (column 6, lines 43-46), and the thus-collected hydrogen sulfide is then reused as a sulfur source for the polymerization reaction. Specifically, in Example 1a of Miyahara et al, H₂S is trapped in the gas absorber (A) charged with the organic amide solvent (NMP). The same process is also employed in the remaining Examples of Miyahara et al. However, Miyahara et al disclose that a significant amount, for example, 25% in Example 1a, of the volatilized hydrogen sulfide is trapped in the distillate (water) collected in the three-necked flask, which is later discharged. One skilled in the art will recognize that the proportion of hydrogen sulfide in the distillate will vary greatly according to changes in conditions of the dehydration step. In addition, it is necessary to exactly measure the amount of hydrogen sulfide contained in the solution in the gas absorber (A) charged with NMP since the molar ratio of the sulfur source to the dihalo-aromatic compound in the reaction cannot be exactly set unless the amount of recycled hydrogen sulfide is exactly measured.

Anticipation under 35 U.S.C. §102 requires that each and every element as set forth in the claims is found, either expressed or inherently described, in a single prior art reference, *In re Robertson*, 49 U.S.P.Q. 2d 1949, 1950 (Fed. Cir. 1999). As discussed in detail above, Miyahara et al fail to teach a production process including a dehydration step in which a volatilized vapor is guided to a linked distillation column, with a fraction taken from the bottom of the distillation

column comprising organic amide solvent which is refluxed into the reaction vessel. Further, Miyahara et al fail to disclose a production process wherein a fraction taken from the top of the distillation column is cooled and a part of the condensed water is refluxed into the distillation column while the remaining water is discharged, and Miyahara et al fail to disclose a production process wherein a predetermined relational expression between the total of the amount of water refluxed into the distillation column and the amount of water discharged without being refluxed, and the amount of hydrogen sulfide discharged from the reaction vessel is used to calculate an amount of discharged hydrogen sulfide, which is then in turn used to calculate the sulfur source remaining in the reaction mixture and to control a charged molar ratio of the sulfur source to a dihalo-aromatic compound. Thus, Miyahara et al fail to describe each and every element as set forth in claim 1. Thus, Miyahara et al do not anticipate claim 1, or any of the claims dependent thereon.

In the Official Action, the Examiner asserted that since Miyahara et al disclose the use of a distillation column, the Miyahara process encompasses the claimed steps and a prepolymer having claimed characteristics could inherently be formed. Applicants respectfully disagree with both of the Examiner's assertions. First, Miyahara et al disclose that the dehydration step is performed by heating the alkali metal sulfide in the organic amide solvent to separate water by distillation outside the reaction system. In contrast, the dehydration step in the process of claim 1 is not outside the reaction system as the distillation column is linked to the reaction vessel and a fraction taken from the bottom of the distillation column is refluxed into the reaction vessel and a fraction taken out of the top of the distillation column is condensed and a portion of the resulting condensed water is refluxed into the distillation column. Thus, even when Miyahara et al employ distillation as mentioned at column 6, line 45, such does not result in a

process as presently claimed. Additionally, Miyahara et al provide no teaching or recognition that the amount of volatilized hydrogen sulfide can be calculated using a predetermined relational expression.

Further, the fact that a certain result or characteristic may occur or be present in the prior art is not sufficient to establish the inherency of that result or characteristic, *In re Rijckaert*, 28 U.S.P.Q. 2d 1955, 1957 (Fed. Cir. 1993). To establish inherency, the extrinsic evidence must make clear that the missing descriptive matter is necessarily present in the thing described in the reference, and that it would be so recognized by persons of ordinary skill; inherency may not be established by probabilities or possibilities and the mere fact that a certain thing may result from a given set of circumstances is not sufficient, *In re Robertson, supra* at 1950-51. In view of the significant differences in the process of claim 1 and the teachings of Miyahara et al discussed in detail above, the evidence of record does not show that the steps of claim 1 are inherent in the Miyahara et al process or that the prepolymer melt viscosity recited in claim 9 is necessarily present in the teachings of Miyahara et al and that it would be so recognized by persons of ordinary skill. Accordingly, there is no basis for asserting that the limitations of any of claims 1-3 and 5-9 are inherent in the teachings of Miyahara et al.

It is therefore submitted that Miyahara et al do not anticipate the process of claim 1 or of any of claims 2, 3 and 5-9 dependent thereon. Similarly, Miyahara et al do not render the presently claimed process use obviously to one of ordinary skill in the art.

More particularly, as discussed above, Miyahara et al disclose a process comprising recovering hydrogen sulfide volatilized out during a dehydration step by causing it to be absorbed in an organic amide solvent outside the system in which the dehydration step is being conducted, and reusing the recovered hydrogen sulfide as a sulfur source in a polymerization

reaction. According to Miyahara et al, the hydrogen sulfide volatilized out in the dehydration step can be recovered and reused, thereby solving various problems resulting from the volatilization of hydrogen sulfide. However, even in Miyahara et al's process, the amount of the volatilized hydrogen sulfide absorbed in the organic amide solvent varies. Accordingly, it is necessary in the production process of Miyahara et al to determine the amount of the hydrogen sulfide volatilized out in the dehydration step and absorbed in the organic amide solvent in order to carefully control the ratio of sulfur source and dihalo-aromatic compound. As noted, such measurements are time consuming and require the use of additional equipment.

On the other hand, the process according to the present invention calculates the amount of discharged hydrogen sulfide as a function of the total of the amount of water refluxed into the distillation column and the amount of water discharged after cooling. These amounts are easily measured and may be done on a continuing basis to accurately control the sulfur source - dihalo-aromatic compound ratio in the production process. As a result, the production process provides a reliable means for producing good-quality poly(arlene sulfides) of consistent melt viscosity.

As the Supreme Court noted in *KSR International Company v. Teleflex Inc.*, 550 U.S. ___, 82 U.S.P.Q. 2d 1385 (2007), rejections for obviousness cannot be sustained by mere conclusory statements; instead, there must be some articulated reasoning or some rationale underpinning to support the legal conclusion of obviousness, 82 U.S.P.Q. 2d at 1396. The Examiner has not provided any articulated reasoning or rationale for one of ordinary skill in the art to modify the various teachings of Miyahara et al to result in the presently claimed production process. In fact, Applicants find no teachings, suggestion or recognition by Miyahara et al that a predetermined relational expression can be determined between discharged hydrogen sulfide and the total of the amount of water refluxed into the distillation column and the amount of water

discharged without being refluxed as recited in claim 1. Thus, Miyahara et al do not render the presently claimed production processes obvious.

It is therefore submitted that the production processes defined by claims 1-3 and 5-9 are neither anticipated by nor rendered obvious over Miyahara et al, and that the rejection under 35 U.S.C. §102(b) or §103(a) has been overcome. Reconsideration is respectfully requested.

It is believed that the above represents a complete response to the Official Action and places the present application in condition for allowance. Reconsideration and an early allowance are requested.

Please charge any fees required in connection with the present communication, or credit any overpayment, to Deposit Account No. 503915.

Respectfully submitted,

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